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RAPID COMMUNICATION

Elliptical micro-ring organic lasers

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Abstract

Multimode laser action was observed from optically excited 2,5-dioctyloxy poly(para-phenylene-vinylene), DOO-PPV, micro-rings coaxially deposited around glass optical fibres of elliptical cross-section. The laser emission was found to be dependent upon the incident angle of the excitation and exhibited linewidths of approximately 1.2 Å, quality factors (Q) exceeding 5000, and thresholds below 0.3 μJ/pulse. Such elliptical organic micro-ring lasers offer increased tailorability in emission properties over more conventional analogues of circular cross-section. Also discussed is the potential for such low-threshold lasers to serve as integrated sources for fibre lasers and amplifiers.

Keywords: polymers and organics, micro-rings, lasers, micro-cavity and micro-disk lasers

Since their first realization over a decade ago [1], organic lasers, particularly in micro-cavity geometries, have enjoyed steady inquiry by both the materials science and optical physics communities [2–7]. This is largely due to the potential for these components to exhibit low-threshold lasing and acceptably high-quality factors, Q, while possessing greater manufacturability at lower cost than inorganic counterparts. In particular, considerable advances have been made in device design to understand and optimize performance and chemistry to control the excited-state dynamics and emission wavelengths and quantum efficiencies. Continued progress towards practical application would be benefited by further enhancing the tailorability of the lasing properties. To this end, this work focused on the fabrication and spectroscopic characterization of organic micro-ring lasers of elliptical cross-section. Such oval micro-cavity lasers are generated by the solution deposition of a light-emissive pi-conjugated polymeric film on an elliptical optical fibre. As is shown, aside from the realization of low thresholds, the lasing spectrum of the elliptical micro-rings was dependent upon the incident angle of the optical excitation. This angle dependence between pump incidence and laser wavelength provides an added degree of control over the performance of micro-cavity lasers.

The pi-conjugated polymer used in this work was 2,5-dioctyloxy poly(para-phenylene-vinylene), DOO-PPV. The DOO-PPV was synthesized following previously reported procedures [8], dissolved in toluene at 70 °C to varying concentrations ranging from 1 to 5 mg ml^{-1} in 1 mg ml^{-1} increments, and doubly filtered through a 0.25 μm PTFE (Teflon) syringe filter in order to remove any suspended or undissolved particles that would lead to optical scattering in the micro-ring.

Micro-rings were fabricated by first taking a 4–5 cm length of the elliptical glass fibre and cleaning its surface in ethanol to remove any dust or particulates on the fibre surface. In a transfer pipette, the DOO-PPV/toluene solution was deposited onto elliptical optical fibres by drip-coating, where a drop of solution is placed onto the glass fibre surface and surface tension is used to yield a smooth uniform coating. The toluene in the solution subsequently evaporates at room temperature, leaving a thin DOO-PPV film of approximately 500 μm in thickness, depending upon the number of drops of solution used.

The elliptical optical fibre was drawn at Clemson University's optical fibre draw laboratories [9]. A cylindrical rod of phosphate glass was ground to have optical flats on two parallel sides. Uncoated glass optical fibre was drawn at approximately 1000 °C, with long and short axes of the substantially elliptical cross-section being approximately 150 and 300 μm, respectively. Surface tension effects during the fibre drawing did lead to some rounding off of the parallel flats. This can be lessened to some degree by a lower-temperature

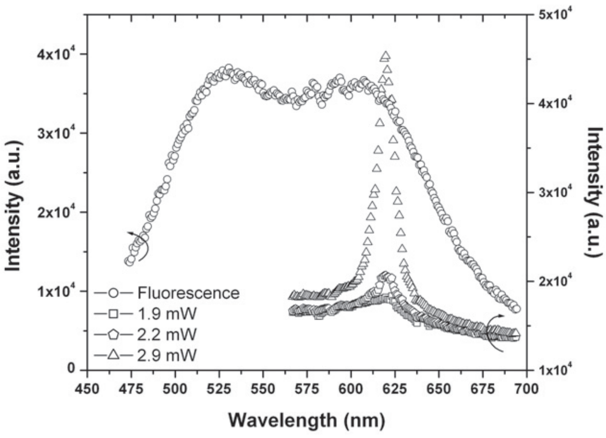

Figure 1. Emission from the DOO-PPV micro-ring as a function of excitation power.

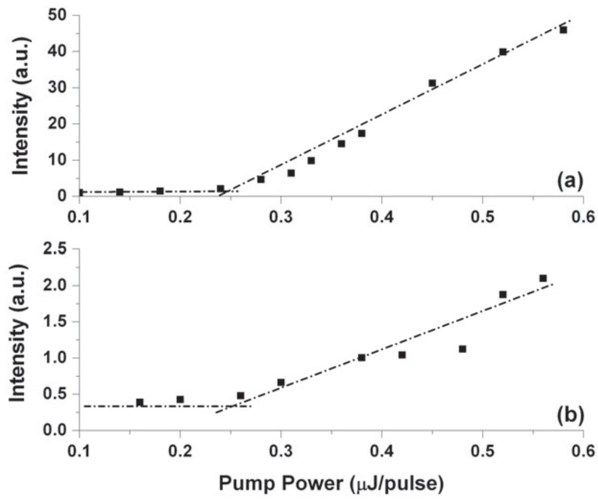

Figure 2. Micro-ring emission intensity as a function of pump power for excitation normal to the minor (a) and major (b) ellipse axes. Dashed lines are only a guide-to-the-eye for approximating the lasing threshold.

draw but, for the proof-of-concept efforts of this work, this ovality was not considered to be consequential to the lasing performance of the micro-rings.

The surface roughness of the micro-rings was characterized by using atomic force microscopy (AFM). The AFM used was a Nanoscope Dimension™ 3100 Atomic Force Microscope (Digital Instruments) operated in the tapping (non-contact) mode.

Photo-excitation of the micro-rings was performed by using a frequency-doubled Nd:YAG regenerative laser amplifier producing 100 ps pulses at 1 kHz repetition rate. The pump beam was focused using a cylindrical lens into a 6 mm × 150 μm stripe, which was applied perpendicular to the micro-ring. Micro-ring samples of 0.1–0.6 mm of lateral length were photo-excited inside a sample holding chamber where vacuum was applied with a continuously running vacuum pump to minimize photo-degradation of the polymer. The emissions from the micro-ring samples were collected from the end of the glass optical fibre and spectrally analysed using a CCD camera with spectral resolution of 0.1 Å. All measurements were made at room temperature and the excitation beam was always perpendicular to the coaxial micro-ring and elliptical optical fibre.

AFM analysis (images not shown) was performed on the micro-ring as a function of DOO-PPV solution concentration. For the films drip-cast from the 2 mg ml^{-1} solution, the root mean square (rms) roughness was measured to be 18.7 nm, whereas the 3 and 5 mg ml^{-1} films exhibited an rms roughness of 20.6 and 40.7 nm, respectively. Accordingly, the higher-concentration solutions yielded rougher surfaces. As relates to subsequent use in a laser oscillator/amplifier system, the surfaces formed from the 2 and 3 mg ml^{-1} solutions are considered to be optically smooth on the basis of the Rayleigh and Fraunhofer criteria.

The Rayleigh criterion defines a surface as being smooth when its roughness is less than $\lambda/8\cos(\theta)$, where λ is the free-space wavelength of the light being used and θ is the incident angle of light to the micro-ring surface, which, for this present consideration, is selected as zero degrees where a maximum in scattering would occur. In the present case, where λ is 630 nm corresponding to the peak emission from DOO-PPV, the surface is smooth up to a maximum value of approximately 79 nm. Similarly, the Fraunhofer criterion defines a surface as being smooth when its roughness is less than $\lambda/32\cos(\theta)$, which here would be below approximately 20 nm. Whereas solutions of all concentrations meet the Rayleigh criterion, only micro-rings made from the 1, 2, and 3 mg ml^{-1} solutions meet the Fraunhofer criterion. Hence, desirous of meeting both conditions in order to minimize any extrinsic scattering from the structures, only micro-rings fabricated from solutions with concentrations of 2–3 mg ml^{-1} were used in the laser investigations.

Figure 1 shows the photoluminescence spectra of the elliptical organic micro-ring as a function of pump energy. The fluorescence is fairly broadband, as is known for DOO-PPV, and exhibited two peaks with maxima at 525 and 625 nm. As the pump energy is increased, the luminescence line narrows as the system becomes more highly excited. For excitation intensities higher than 0.3 μJ/pulse, multimode laser emissions were observed. As is shown in figure 2, the lasing characteristics of the micro-ring depended on whether the excitation was normal to the ellipses' major or minor axis.

The exact threshold for lasing is critical to the way the sample was prepared and the experiment was carried out. The typical laser threshold observed was in the range of 0.15–0.5 μJ/pulse (1.5–5 mJ cm^{-2}). These values are comparable to the reported values for dye doped lasers [1] and also correspond approximately to the threshold observed in a cylindrical micro-cavity [10].

The above-noted angular threshold dependence suggested that other laser parameters might also be affected by the angle of the pump beam with respect to the micro-ring axes. Accordingly, the laser spectrum was analysed at a series of angles relative to the major axis. As is shown in figure 3, the multimode nature of the laser was preserved and there was a noticeable shift in the spectral envelope of the lasing modes shifting to shorter wavelengths as the angle of excitation was rotated towards the minor (short) axis of the ellipse. The

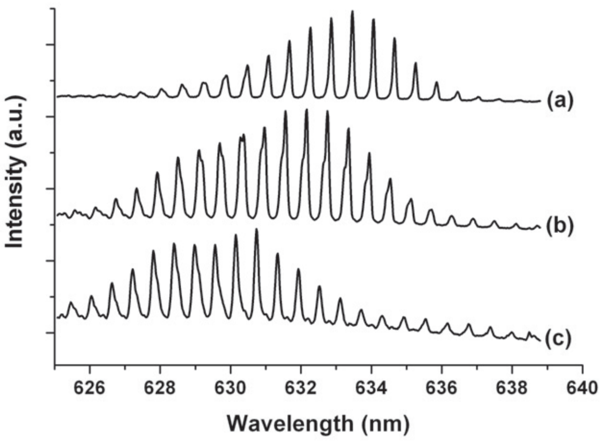

Figure 3. Emission from the DOO-PPV micro-ring as a function of angle of excitation with respect to the fibre major axis: (a) 10°, (b) 30°, and (c) 50°.

five (5) most intense peaks in each spectrum was evaluated and averaged yielding linewidths of approximately 1.2 Å and quality factors, Q, exceeding 5000.

It was noted in the experimental section that the micro-ring emissions were collected from the end of the glass optical fibre and analysed. This is unexpected and the practical importance of this should not be overlooked. The ability to collect the micro-ring emissions from the end of the substrate optical fibre implies that the micro-ring modes couple to the propagating modes of the elliptical bulk glass fibre. This is a departure from using a fibre of circular cross-section as the substrate for the micro-rings. In that case, the orthogonality between the micro-ring lasing modes and the propagating modes of the circular fibre preclude couplings, unless there is some perturbation to break this degeneracy [11, 12]. The ability to couple the micro-ring laser emission directly into the propagating mode of the fibre may permit a novel approach to low-threshold pumping of active optical fibres and fibre lasers [13]. In this case, inexpensive broad-area pump diodes of reduced beam quality could be used to excite the micro-ring whose high-Q laser emissions then couple into an active fibre yielding excitation of the doped core. The micro-ring's excitation angle-dependent laser emission could be used as a further way to tailor the excitation to optimize absorption by the dopant. Lastly, since these micro-rings are easily deposited on the glass fibre, it is certainly possible during the fiberization process (just like the protective polymer coatings) that long lengths of the integrated micro-ring pump sources (i.e. moving from micro-rings to polymeric laser coatings) may be generated to provide a distributed excitation at lower power levels than by direct end-pumping.

Micro-rings of the pi-conjugated polymer DOO-PPV were deposited onto glass optical fibres of elliptical cross-section. Upon optical excitation, laser emissions were observed from the organic micro-ring above a threshold of approximately 0.3 μJ/pulse. The micro-ring laser output exhibited linewidths of approximately 1.2 Å and quality factors of above 5000. The spectral peak position of the lasing modes was observed to be a function of the angle of excitation with respect to the ellipse axes, which can be used to fine-tune the laser wavelengths. Micro-ring laser emissions were found to couple into the propagation modes of the glass optical fibre due to its ellipticity, which breaks the symmetry associated with the orthogonality of propagating modes between the micro-ring and fibre. This may permit a novel way to convert a low-quality pump sources into more efficient excitation of an active fibre laser or amplifier.

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